

# A spatial refractive index sensor using whispering gallery modes in an optically trapped microsphere

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**We propose the use of an optically trapped, dye doped polystyrene microsphere for spatial probing of the refractive index at any position in a fluid. We demonstrate the use of the dye embedded in the microsphere as an internal broadband excitation source, thus eliminating the need for a tunable excitation source. We measured the full width at half maximum of the TE and TM resonances, and their frequency spacing as a function of the refractive index of the immersion fluid. From these relations we obtained an absolute sensitivity of  $5 \times 10^{-4}$  in local refractive index, even when the exact size of the microsphere was not *a priori* known.**

Microspheres can act as high Q resonators in the optical regime<sup>1,2</sup> and can store electromagnetic waves by repeated total internal reflection. Constructive interference results in an enhanced internal field which is called a whispering gallery mode (WGM). Because of the evanescent interaction between the WGM and the surrounding medium the WGM's resonance frequency is sensitive to changes in the refractive index of the sphere's surroundings. This sensitivity makes microspheres an useful tool in sensory applications. Recently Hanumegowda *et al.* demonstrated the use of microspheres in refractometric sensing.<sup>3</sup> Moreover Vollmer *et al.*<sup>4</sup> and White *et al.*<sup>5</sup> demonstrated protein detection using microspheres. In all these studies<sup>3-5</sup> narrowband light from a tunable diode laser was evanescently coupled to the microsphere via an optical fiber. Spatial mapping of the refractive index was not possible because the microsphere could not be freely moved through the sample.

In this letter we demonstrate the use of a dye-doped polystyrene microsphere as a refractometric sensor. The polystyrene microsphere was trapped with optical tweezers, by which it could be positioned anywhere within the fluid. It can therefore be used to sense the refractive index in spatially inhomogeneous media, for example near interfaces or in mixing flows. In this system we did not need a tunable laser to excite WGMs, instead we used the dye embedded in the microsphere itself as a broadband excitation source. The absolute frequency of the WGM, which was used to probe the refractive index in previous studies,<sup>3-5</sup> is very sensitive to the size of the sphere. Microspheres synthesized in bulk always have a considerable size polydispersity, and therefore a wide distribution of absolute resonance frequencies. Measurements of the absolute resonance frequency of a particle that has not been individually calibrated does not provide information about the refractive information of the medium. In contrast, we show that both the full width at half maximum (FWHM) of the emission peaks and the frequency spacing between adjacent transverse electric

(TE) and transverse magnetic (TM) modes are sensitive and robust probes of the refractive index of the sphere's surroundings. We show that a calibration of the individual microspheres is not needed in this case.

The FWHM of a WGM is determined by the loss upon total internal reflection at the curved surface of the microsphere. These losses depend on the mode number  $l$  and the refractive index contrast  $m = m_0/m_I$ , where  $m_0$  and  $m_I$  are the refractive indices of the immersion medium and the microsphere respectively. The FWHM of a WGM can be expressed as<sup>7</sup>

$$\frac{\Gamma}{2} = [Nx^2n_l(x)^2]^{-1}, \quad (1)$$

with  $n_l$  the spherical Neumann function,  $x = m_0ka$  the size parameter at which the WGM occurs, where  $k$  is the wave vector in vacuum and  $a$  the radius of the microsphere, and

$$N = \begin{cases} m^2 - 1 & \text{for TE modes} \\ (m^2 - 1)[\mu^2 + (\mu^2/m^2 - 1)] & \text{for TM modes,} \end{cases} \quad (2)$$

where  $\mu = \nu/x$ , with  $\nu = l + 1/2$ .

The frequency spacing between a TE and TM mode of the same mode number is due to a difference in phase shift of s and p polarized light upon reflection at the sphere surface. The absolute frequency at which a WGM occurs is defined as the real part of the pole of the scattering coefficient,<sup>6</sup> and we calculated the frequency spacing by numerically calculating the poles for TE and TM modes.

In our experiments we used a suspension of dye-doped polystyrene microspheres in water (G1000, Duke Scientific Corporation). The manufacturer specified a mean radius of  $5.0 \pm 0.3 \mu\text{m}$  (standard deviation). We measured the size distribution of 24 microspheres and found a mean radius of  $5.3 \mu\text{m}$  with a maximum deviation of only 0.5 %. Taking into account a conservative error margin, we assumed that the size of any sphere we trap

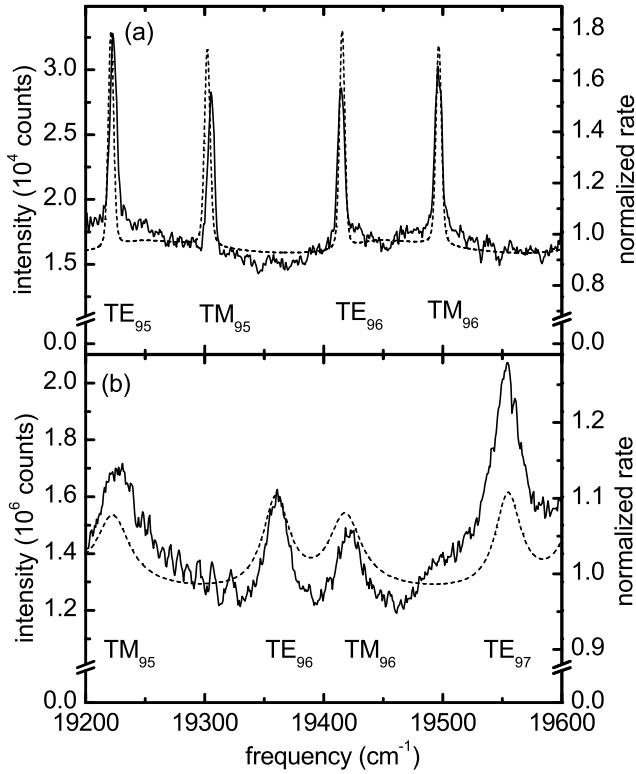


Figure 1: Measured emission intensity (solid lines) of single dye doped polystyrene microspheres immersed in (a) water ( $m_0 = 1.3325$ ) and (b) a mixture of water and ethylene glycol ( $m_0 = 1.3978$ ). The dashed lines show the normalized radiation rates as predicted by EMT for (a)  $a=5.33 \mu\text{m}$ , and (b)  $a=5.319 \mu\text{m}$ .

was  $5.30 \pm 0.05 \mu\text{m}$ . The microspheres were doped with 2% dye which emits in the green part of the spectrum ( $\lambda = 480 - 540 \text{ nm}$ ). The sample holder was a borosilicate glass capillary with inner dimensions  $0.1 \times 2 \times 40 \text{ mm}^3$ .

With optical tweezers operating at  $1064 \text{ nm}$  we held a single microsphere far away from any surface. The excitation light source was a continuous wave Argon laser (Spectra Physics Satellite 2016) emitting at  $488 \text{ nm}$  with a maximum output power of  $30 \text{ mW}$ , attenuated to  $500 \mu\text{W}$  on the sample. The laser light was slightly defocused on the sphere, so that the whole sphere was illuminated. The emission of the dye was collected through a water immersed objective (NA 1.2) and detected by an ICCD (Princeton Instruments ICCD 576), connected to a spectrograph (Oriel Instruments, MS257) which has a Gaussian response function with a width of  $4.5 \text{ cm}^{-1}$ .

Tuning of the refractive index of the immersion fluid  $m_0$  was achieved by mixing water with ethylene glycol ( $m_0 = 1.4317$  at  $293 \text{ K}$  and  $589 \text{ nm}$ ) in various fractions. Ethylene glycol does not chemically affect polystyrene and is miscible with water in all volume fractions. Refractive indices of the mixtures were measured with an Abbe refractometer type 1T which is temperature stabilized and has an accuracy of  $2 \times 10^{-4}$  in refractive index.

We compared our experimental emission spectra to an

elaboration of Mie theory (EMT) developed by Chew *et al.* describing the emission of dipoles inside a spherical cavity.<sup>8,9</sup> The only parameters were the refractive index of the sphere  $m_I$ , the refractive index of the immersion medium  $m_0$ , and the sphere radius  $a$ . The refractive index of the sphere was measured to be  $m_I = 1.586 - i \cdot 1.5 \times 10^{-5}$ .<sup>10</sup> The refractive index of the immersion fluid  $m_0$  was measured with the refractometer. The only free parameter in our system was the radius of the sphere  $a$ . The theoretical emission spectra were corrected for the response function of the spectrograph.

In Fig. 1 we show the emission spectra obtained from a single dye doped polystyrene microsphere immersed in (a) water ( $m_0 = 1.3325$ ) and (b) a mixture of water and ethylene glycol ( $m_0 = 1.3978$ ). Sharp peaks were observed in the spectrum due to WGMs. The dashed lines show the radiation rates (normalized to the rate in bulk polystyrene) as predicted by EMT for (a)  $a = 5.33 \mu\text{m}$ , and (b)  $a = 5.319 \mu\text{m}$ . Comparing the spectra in (a) and (b) we observed that the decrease in refractive index contrast induced an increase in FWHM of the WGMs and a decrease in the spacing between the adjacent TE and TM modes.

As we have shown in Fig. 1 it is possible to obtain  $m_0$  and  $a$  by fitting the experimental spectra with a theoretical emission spectrum. However, the initial values for the parameters must be chosen rather carefully. The same parameters can be obtained in a more straightforward way by comparing only the FWHM and the  $\text{TE}_{96}$ - $\text{TM}_{96}$  mode spacing to predictions from EMT. Both the FWHM and the mode spacing were determined by fitting a Voigt profile<sup>11</sup> to an individual WGM in the measured emission spectrum. The Voigt profile models the convolution of the Lorentzian width of the resonance and the Gaussian response function of the spectrograph. To accurately fit the resonance shape, we subtracted a quadratic fit of the background intensity.

In Fig. 2 we show the Lorentzian FWHM as a function of  $m_0$  for the  $\text{TE}_{96}$  mode. The size of the error bars was taken from the error in the Lorentzian width of the fits. The large error at  $m_0 = 1.41$  was due to a low signal to noise ratio resulting from a low refractive index contrast. The theoretical FWHMs (solid lines) were calculated for the biggest and smallest spheres in the measured size distribution, and were calculated from Eq. (1). It can be seen that the FWHM is very insensitive to the sphere size as the calculated curves are almost identical.

In Fig. 3 we show the experimental  $\text{TE}_{96}$ - $\text{TM}_{96}$  mode spacing, obtained from the central frequency of the fitted Voigt line shapes. The error bars are the uncertainty in the central frequency of the Lorentzian fits. The theoretical mode spacing is again calculated for the biggest and smallest spheres in the measured size distribution, and the result is shown as a grey area. It can be seen that the  $\text{TE}_{96}$ - $\text{TM}_{96}$  mode spacing is slightly more sensitive to sphere size than the FWHM (Fig.2).

Figs. 2 and 3 show that our measurements correspond very well to the theoretical model, even though the size of the trapped microsphere varies between measurements

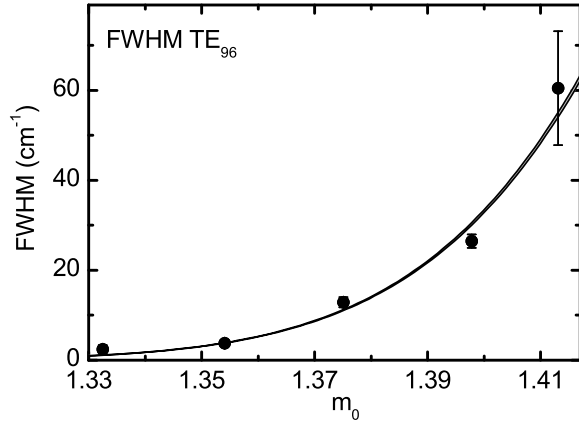


Figure 2: Measured Lorentzian FWHM versus  $m_0$  for the  $TE_{96}$  mode. The solid lines show the theoretical FWHM for the biggest and the smallest spheres in the measured size distribution, and was calculated from Eq. (1).

due to size polydispersity. The polydispersity in sphere size introduced an inaccuracy in the theoretical values for the FWHM and TE-TM mode spacing of  $< 1\%$ .

One can use either the FWHM or the mode spacing to measure the refractive index of the medium. However, the sensitivity and robustness of these methods is not the same. The  $TE_{96}$ - $TM_{96}$  mode spacing is nearly linear in our refractive index range with a slope of  $385\text{ cm}^{-1}$  per refractive index unit. An accuracy in width of  $2\text{ cm}^{-1}$  was easily achieved for refractive indices below 1.39, resulting in a sensitivity of nearly  $5 \times 10^{-3}$  in refractive index. When measuring the FWHM of a WGM the sensitivity of the sensor is higher, and varies throughout our refractive index domain. From the slope of the curve in Fig. 2 we obtained a sensitivity of  $5 \times 10^{-4}$  in refractive index around  $m_0 = 1.41$ . The FWHM method leads to the most precise measurements in the ideal case, however, this method is very sensitive to contamination at the sphere surface. The mode spacing is a more robust method and may therefore be used when contamination cannot be excluded.

In conclusion we demonstrated a spatial probe for the refractive index comprising an optically trapped dye doped microsphere. The use of optical tweezers gives us the freedom to measure the refractive index anywhere in the fluid. This freedom allows for spatial mapping of refractive index gradients or inhomogeneities caused by, for example, flow in microfluidic devices or chemical processes. The emission of an embedded dye acts as the internal source to excite the WGMs in the microsphere. We measured the FWHM and the frequency spacing between TE and TM modes. The measured values show excellent agreement with theoretical predictions, confirming the suitability of the system for spatial probing of the refractive index without the need for individual calibration. Very recently, Knöner *et al.*<sup>12</sup> have demonstrated a method to determine the refractive index of a trapped microsphere by measuring the trapping force. Their and

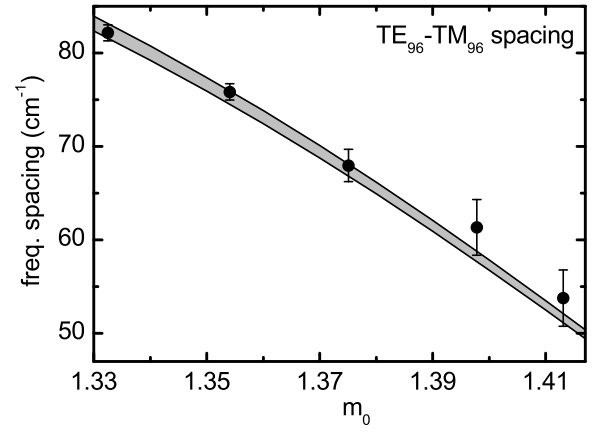


Figure 3: Measured  $TE_{96}$ - $TM_{96}$  mode spacing versus  $m_0$ . The grey area shows the theoretical mode spacing calculated for the biggest and smallest spheres in the measured size distribution.

our methods are complimentary and could easily be combined, providing a versatile system for sensory applications.

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